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CONCEPTS OF IDEAL AND NONIDEAL EXPLOSIVES.(U)

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CONCEPTS OF IDEAL AND NONIDEAL EXPLOSIVES

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US ARMY ARMAMENT RESEARCH AND DEVELOPMENT COMMAND
LARGE CALIBER
WEAPON SYSTEMS LABORATORY
DOVER, NEW JERSEY

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<table border="0"> <tr> <td>Ideal explosives</td> <td>Thermohydrodynamic theory</td> </tr> <tr> <td>Nonideal explosives</td> <td>Equations of state</td> </tr> <tr> <td>Diameter effect</td> <td>Ammonium nitrate</td> </tr> </table>			Ideal explosives	Thermohydrodynamic theory	Nonideal explosives	Equations of state	Diameter effect	Ammonium nitrate
Ideal explosives	Thermohydrodynamic theory							
Nonideal explosives	Equations of state							
Diameter effect	Ammonium nitrate							
20. ABSTRACT (Continue on reverse side if necessary and identify by block number)								
<p>The purpose of this report is to stimulate discussion on the nonideality of ammonium nitrate and its composite explosives. The concept of ideal and non-ideal explosives is explored, which leads to a discussion of the relationship between theory and experiment of the diameter effect. The behavior of some nonideal explosives is considered, and it is suggested that these explosives may tend to be ideal at large diameters. For composites containing nonideal</p>								

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20. ABSTRACT (cont)

explosives, the trend in approaching ideal behavior may be due, in part, to the lowering of the large diameter requirement by the small particle size and the relatively low effective density of the nonideal explosive in the composite.

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INTRODUCTION

Because of its obvious advantages of cheapness and calculated performance, ammonium nitrate (AN) has become an important military consideration in recent times. Unfortunately, one of its limiting disadvantages is its nonideal behavior. Therefore, the main thrust in utilizing AN for military purposes consists of formulating intimate mixtures of AN and various fuels as a means of approaching ideal behavior. This assumes that we have an adequately functioning concept of ideal behavior to serve as a criterion. It is this concept and its application to non-ideal explosives, particularly AN, that will be examined with the purpose of stimulating discussion on the subject.

IDEAL AND NONIDEAL EXPLOSIVES

It is generally accepted that an ideal explosive is one in which the detonation velocity corresponds to the theoretical maximum detonation velocity calculated by the thermohydrodynamic theory (ref 1). The detonation velocity of an explosive is determined experimentally from the steady value obtained at a sufficiently long distance from the initiator and of such charge diameter that no further increase in length or diameter will increase the detonation velocity. A nonideal explosive is one in which the observed detonation velocity is lower than the calculated ideal value. Nonideal detonation behavior can occur for all explosives when the charge diameter is less than the minimum diameter for ideal detonation and greater than the critical or failure diameter. Actually, the definition of ideal and nonideal should also include the C-J pressure and expansion isentrope, as well as the detonation velocity. Unfortunately, there are relatively few measurements of detonation properties other than velocity, and many of those that have been made are of questionable accuracy because of the experimental difficulties and differing interpretations of the same data (ref 2). Since the detonation velocity is a more easily determined quantity, most tests of ideality-nonideality refer to it only. However, it is the least sensitive of the detonation parameters.

DIAMETER EFFECT

The concept of ideality-nonideality involves the diameter of the charge. In order to obtain some idea of the reaction zone length as well as the infinite-diameter velocity at a given density, it has been customary to linearly extrapolate detonation velocity-reciprocal diameter data according to the approximation of Eyring's theory,

$$\frac{D}{D^*} = 1 - \frac{a}{d}, \quad (1)$$

where D is the detonation velocity, D^* is the infinite diameter detonation velocity, d is the diameter of the charge, and a is the reaction zone length (ref 3). However, this equation is really an empirical one, as Eyring pointed out. From a dimensional analysis alone,

$$\frac{D}{D^*} = f\left(\frac{a}{d}\right). \quad (2)$$

Expanding, with the boundary conditions that $\frac{D}{D^*} = 1$ when $\left(\frac{a}{d}\right) = 0$, gives

$$\frac{D}{D^*} = 1 + A\left(\frac{a}{d}\right) + B\left(\frac{a}{d}\right)^2 + \dots, \quad (3)$$

so that the linear equation is valid only for small values of $\left(\frac{a}{d}\right)$ or for values of D close to D^* (ref 4). In addition, Eyring applied the equation to unconfined explosives, and obtained other equations for confined explosives. There is yet no quantitative theory that can relate unconfined and confined explosives (ref 5). In general, then, a plot of D versus the reciprocal diameter or radius is not linear, and this is shown by Cambell and Engelke (ref 6) who fitted data to the equation

$$\frac{D}{D^*} = 1 - \frac{A}{(R-R_c)} \quad (4)$$

where R is the radius of the charge, and A and R_c are length parameters. It can be seen that as $R_c \rightarrow 0$, the equation reduces to Eyring's linear equation. Figure 1 shows some of the results. Here, for the most part, only those detonation velocities close to D^* can be fit to the linear equation, and deviations will become important at different radii for each explosive. For example, with XTX-8003, extremely small radii are required before nonlinearity is observed. On the other extreme, Amatex/20 would give an incorrect D^* extrapolation if measurements were made with diameters less than about 1 1/2 inches. The situation is not a great deal better with cast and creamed 2,4,6-trinitrotoluene (TNT).

THERMOHYDRODYNAMIC THEORY

General Assumptions

The thermohydrodynamic theory involves the following general assumptions:

1. One-dimensional flow.
2. Infinite reaction rate, which implies an infinitely-small reaction zone length.
3. Chemical equilibrium at the C-J plane.
4. C-J condition: The particle velocity at the C-J plane is sonic with respect to the detonation front.

Since the assumptions are not valid for real explosives, it is remarkable that the theory can be applied to a wide variety of explosives with reasonable success. It appears that the theory is not too sensitive to the assumptions.

Equations of State

In order to apply the theory to calculation of detonation properties of explosives, an equation of state for the detonation products is required. The Encyclopedia of Explosives (ref 7) lists more than 50 of them. The equation of state that is now used most often is the Becker-Kistiakowsky-Wilson (BKW) (ref 8). Other equations of state that are sometimes used include the Jacobs-Cowperthwaite-Zwisler (JCZ-3) (ref 9) and the Lennard-Jones-Devonshire (LJD) (ref 10). Each has its advantages. For example, the JCZ-3 gives better expansion isentropes than the BKW, but not quite as good agreement with detonation velocity and C-J pressure as does the BKW.

When applied to real explosives, the equations of state must be calibrated with well-known detonation values. Various sets of parameters were used in the BKW equation of state, but when more experimental detonation values became available, Mader (ref 8) attempted to improve agreement, but found that one set of parameters for the BKW equation of state could not give satisfactory agreement between theory and experiment. Therefore, two sets were used, one set calibrated with 1,3,5-trinitro-1,3,5-triazacyclohexane (RDX) and another with TNT. In this way, one set could be used for explosives having large quantities of carbon in the products, such as with TNT and 1,3,5-triamino-2,4,6-trinitrobenzene (TATB), and the other set for other explosives, such as pentaerythritol tetranitrate (PETN), RDX, and 1,3,5,7-tetranitro-1,3,5,7-tetrazacyclooctane (HMX).

Because of the way the equations of state are calibrated, it is generally assumed that they apply to CHNO explosives only. Nevertheless, application to other explosives have generally given reasonable results. In order to further improve the results, Finger and his coworkers (ref 11) at Lawrence Livermore Laboratory (LLL) have reparameterized the BKW equation of state with a wider set of explosives. HNO, CNO, and fluorine-containing compounds, as well as CHNO explosives, were used. This single set of parameters gives improved results over a wide range of explosives, but does not generally give quite as good results as with the double set of parameters for CHNO explosives. The reparameterized BKW is designated as BKWR.

Application to AN

The thermohydrodynamic theory as incorporated in the TIGER-BKW code has been used to calculate the detonation properties of ideal explosives, but it may be questioned whether the code would be applicable to nonideal explosives. Nevertheless, calculations were made which formed the basis for discriminating between ideal and nonideal explosives, and led to efforts to improve the performance of a cheap nonideal explosive, AN, by the use of additives. But, under some conditions, even with ideal explosives, the detonation properties cannot be accounted for by the thermohydrodynamic theory. For example, in the detonation velocity-density plot, TNT undergoes a sharp change of slope at a density of 1.55 g/cm³ (ref 13). Ammonium nitrate is generally considered to be an explosive whose nonideal behavior is significant. Thermohydrodynamic calculations of its detonation properties have been made for some time. Some of these are shown in table 1.

Table 1. Calculated ideal properties of AN at $\rho_0 = 1.725 \text{ g/cm}^3$

	Taylor's method (ref 14, 15)	KSM (ref 16)	TIGER BKW	TIGER BKWR	LJD (ref 17)
D (mm/sec)	6.0	6.49	7.84	7.43	7.68
P_{CJ} (kbar)		182	210	190	217
T ($^{\circ}\text{K}$)			470	1177	1120
γ		2.99	3.97	3.94	3.69

Here the detonation velocity calculated by the Kamlet Simple Method (KSM) (ref 16) is relatively close to that calculated by Evans (ref 14) using Taylor's method described in his book (ref 15). The KSM values, which were determined with N_2 , H_2O and O_2 as products, are only slightly less than those for TNT. The TIGER and LJD calculations (ref 17) are somewhat higher than for TNT. We can immediately observe that the temperatures are unusually low. The higher BKWR temperature is due to the fact that it was designed to give higher temperatures than those obtained with the BKW. In general, for most other explosives, temperatures calculated with the BKW are probably too low and those with the LJD are probably too high. Unfortunately, there have been too few temperature measurements made on detonating explosives. Although, for the most part, there is relatively good agreement between calculated and measured temperatures, where available, the equations of state were not calibrated to produce experimental temperatures. However, in those cases where they do not agree, the disagreement does not generally influence the calculations of the detonation velocity and pressure. We can also observe that the TIGER and LJD calculated values for the adiabatic exponent, γ , are extremely large. The value of 2.99 obtained with KSM is one that would be expected for ideal explosives at the same density and can also be calculated for the AN density with Kamlet's "Rule for Gamma" (Ref. 18). A large γ indicates a very steep isentrope, and consequently a good explosive for fragmentation, but a poor one for accelerating metal. Since γ is dependent upon the values of the detonation velocity and pressure, the large value for γ questions the validity of the calculations. In order to throw some light on the question, let us look at explosives similar to AN and a method of classifying explosives by their experimental behavior rather than by the thermohydrodynamic theory.

GROUP 1 AND GROUP 2 EXPLOSIVES

Price (ref 5) observed that explosives could be divided into two groups. In figures 2a and 2b, we have critical diameter - % TMD plots. Here, Group 1 explosives are those whose critical diameters decrease with increasing density. Examples are most of the explosives that we have been calling ideal, such as TNT and RDX. Group 2 explosives are those whose critical diameters increase with increasing den-

sity. Most explosives in this group consist of oxidizer - fuel mixtures, but also include some pure explosives such as AN, AP, and HN. It may appear that Group 2 explosives are the nonideal ones, but as we shall see, HN is an ideal explosive as defined originally. Figures 3a and 3b show the effect of particle size on the critical diameter for both groups. In both groups, the critical diameters are lowered by the fine particles. When the detonation velocity of the two groups are plotted against % theoretical maximum density for various charge diameters, as shown in figures 4a and 4b, it can be seen that for the Group 1 explosives, ideal behavior is approached at all diameters at the theoretical maximum density. On the other hand, Group 2 explosives show entirely different behavior. Here, ideal behavior is approached at lower densities, and all diameters appear to come to a point somewhere to the left of the curve. Extrapolation of D versus reciprocal diameter data to the right of the peaks in the nonlinear region, would give incorrect ideal detonation velocities. The ideal line on the curve was obtained by extrapolating the data to the left of the peaks, which are linear. Figure 5 shows the same Group 2 behavior of HN (ref 19). We can also see the lower density data performed in glass do not seem to show a diameter effect. However, the data are significantly above the high density extrapolation of the ideal behavior data. This may have been caused by the difficulty of determining accurate densities in this low density region and the moisture pickup of this hygroscopic material, which is more important at the lower densities. The experimental results are compared with the older Ruby code (ref 20) and with TIGER-BKWR code calculations. The latter, as expected, gives somewhat better results at the higher densities since it was calibrated for this type of HNO explosive. However, the slopes of both calculations do not appear to be correct. Nevertheless, since the calculations are in reasonable agreement with the more accurate experimental results at the larger diameters and densities, which are of most interest, HN is considered an ideal explosive. Yet, HN has a somewhat higher γ than for normal explosives, although not as large as that for AN.

AN AND AN COMPOSITES

Ammonium nitrate is expected to show typical Group 2 explosive behavior. Unfortunately, the reaction zone length of AN is so large that experiments of significantly large diameters and densities are not feasible. If you must have a number, the failure diameter of AN has been estimated from a model by Evans as 6600 km (ref 14). Since Group 2 explosives approach ideal behavior at the lower densities, it should be possible to study ANFO, which is AN with 6% fuel oil, to improve the oxygen balance, and has a density of 0.8-1.0 g/cm³. It has been observed that ANFO showed ideal performance at sufficiently large diameters, at least 15 inches (ref 17,21). With all the considerations presented, it is reasonable to expect that at sufficiently large diameters, AN should show ideal behavior. Therefore, the difference between an ideal and a nonideal explosive may not be a difference in kind, but one of degree. The important parameter is the ratio of the diameter to the reaction zone length. It also appears that in some of its aspects, such as the detonation velocity and C-J pressure, the thermohydrodynamic theory may be applicable to AN.

The behavior of AN at low densities coupled with small particle size can be utilized to produce explosives approaching ideal behavior by the use of ideal explo-

sive additives. This would lower the large diameter requirement. The best composite, from the small particle consideration, would be one that forms a solid solution. Lacking this, the next best composite would be a eutectic mixture. However, as Askt and Hershkowitz have shown, the manner in which this two-phase mixture is prepared makes a significant difference (ref 22). As examples in the approach of AN to ideal behavior are the well-known AN/EDDN (ref 22) and AN/ADNT (ref 23) eutectics. In table 2, the eutectics of these two systems are considered.

Table 2. AN/EDDN and AN/ADNT eutectics (refs 22, 23)

Eutectic composite	ρ_o (g/gm ³)	Mole fraction of AN	Volume fraction of AN	Effective ρ_o of AN
AN/EDDN	1.65	0.68	0.46	0.79
AN/ADNT	1.67	0.58	0.37	0.65

For explosives in a constant volume shell, the volume fraction is more significant than the mole fraction. From the volume fraction we can obtain the effective density of AN in the composites. Here, we can see that it is the low effective densities of the AN component that lowers the diameter requirement to ideal behavior. However, there are other factors operating that are little understood, such as the rate of reaction between the components in a detonation (ref 24). It is only in understanding some of these factors that ideal explosives having high effective AN densities coupled with low diameter requirements can be produced.

CONCLUSIONS

The concepts of ideal and nonideal explosives depend upon the charge diameter. In comparing thermohydrodynamic calculations with experimental values, it is absolutely necessary that the measured values represent those at sufficiently large charge diameters.

Extrapolation of D versus 1/d data may give incorrect results depending upon the nature of the explosive.

Although there are exceptions, thermohydrodynamic theory based in such codes as TIGER-BKWR, can calculate some aspects of detonation behavior of a wide variety of explosives, probably including AN, with reasonable accuracy. However, there is a great deal of room for improvement as our measurements and understanding become more sophisticated.

In general, explosives may be classified into two groups depending upon their critical diameter versus density plots.

The approach to ideality of AN composites is facilitated by the low effective density of the AN and the fine particle sizes produced by eutectics. However, other factors, such as lack of complete interaction between components, can degrade performance.

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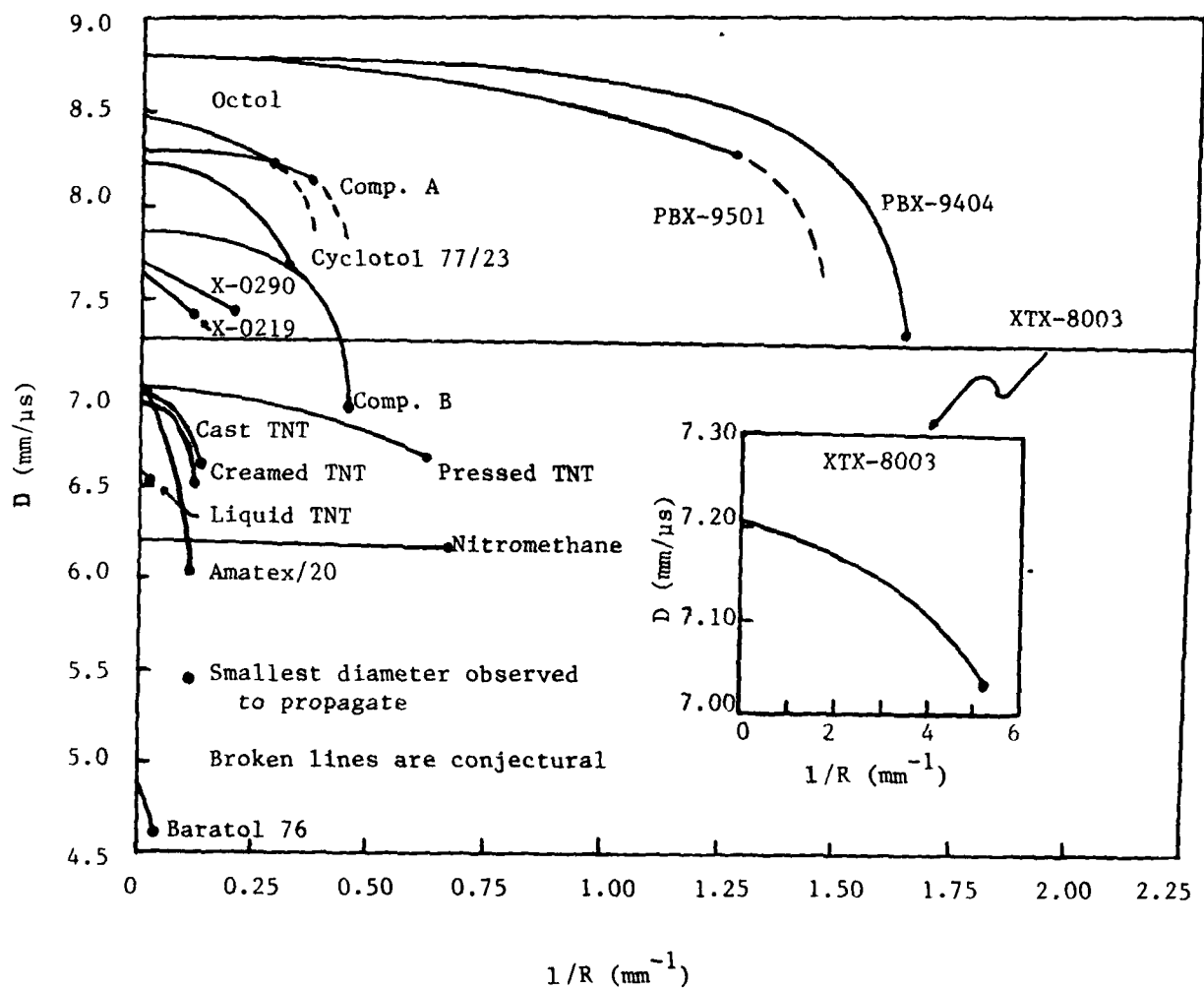
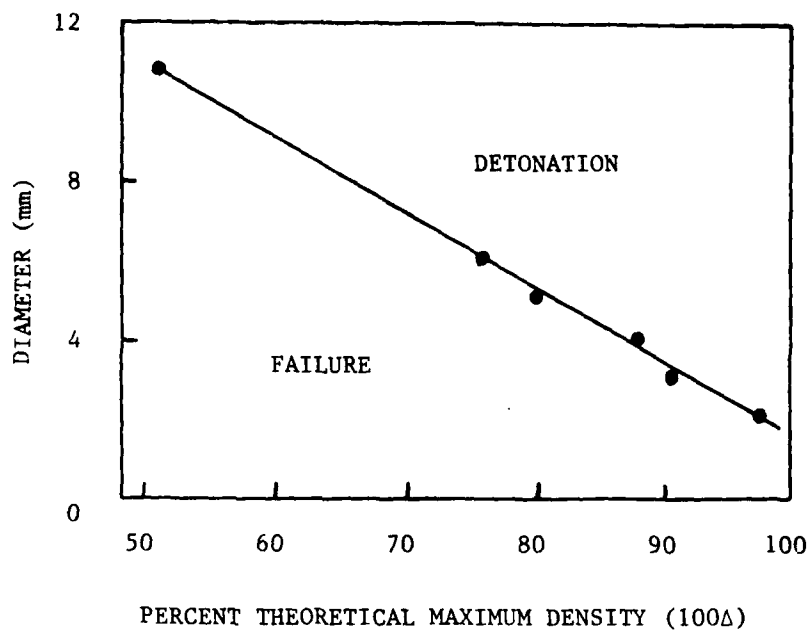
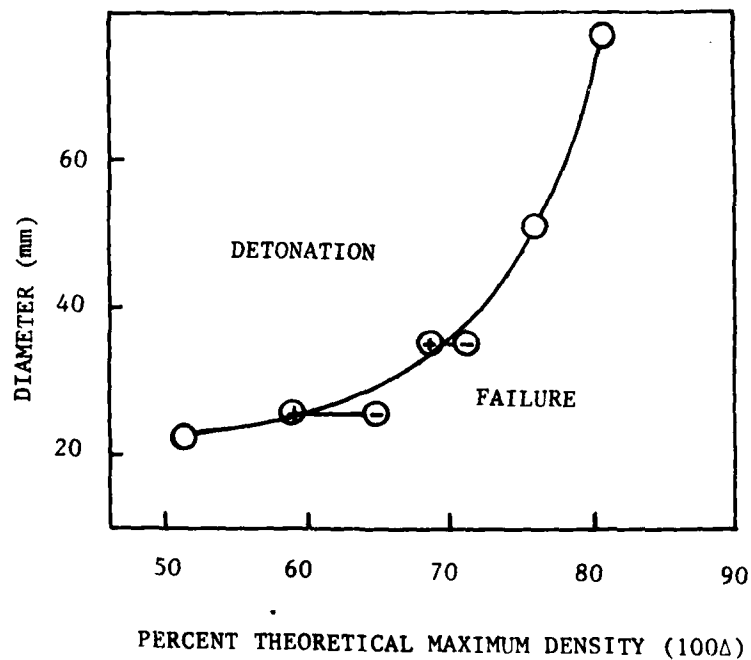


Figure 1. Composite of diameter-effect curves in the D -versus- $1/R$ plane (ref 6)

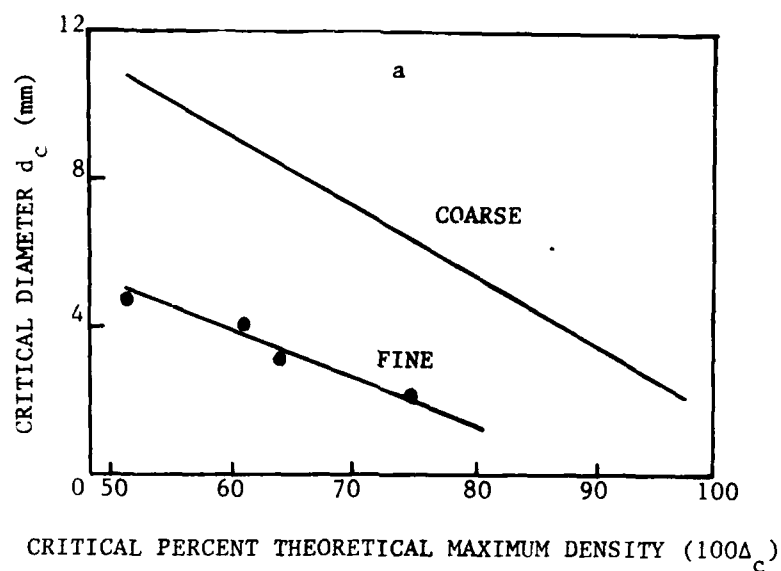


(a) - Group 1 - Example shown is TNT (grain size 0.07 to 0.2 mm)

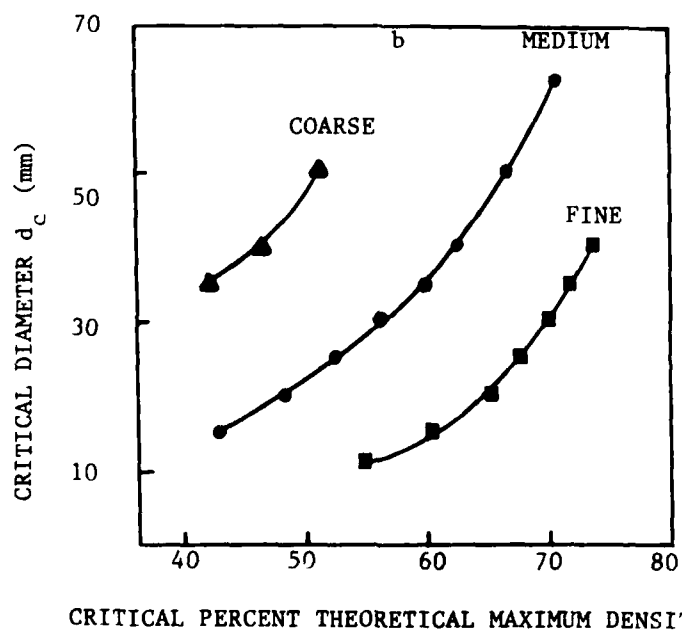


(b) - Group 2 - Example shown is AP (average particle size 10 μ)

Figure 2. Detonability limits in the charge diameter-porosity plane (ref 5)

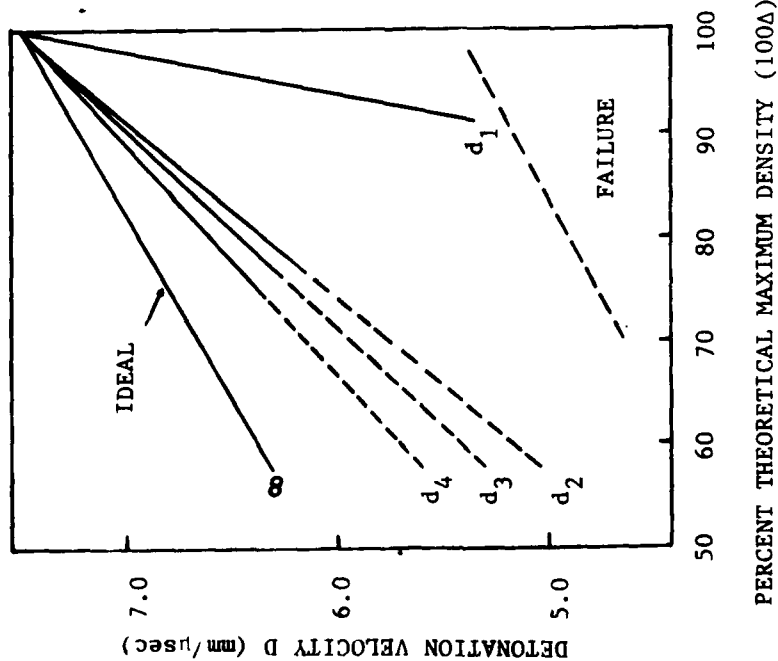


(a) Group 1 - Example shown is TNT. Grain sizes are 0.07 to 0.2 mm and 0.01 to 0.05 mm for coarse and fine, respectively.

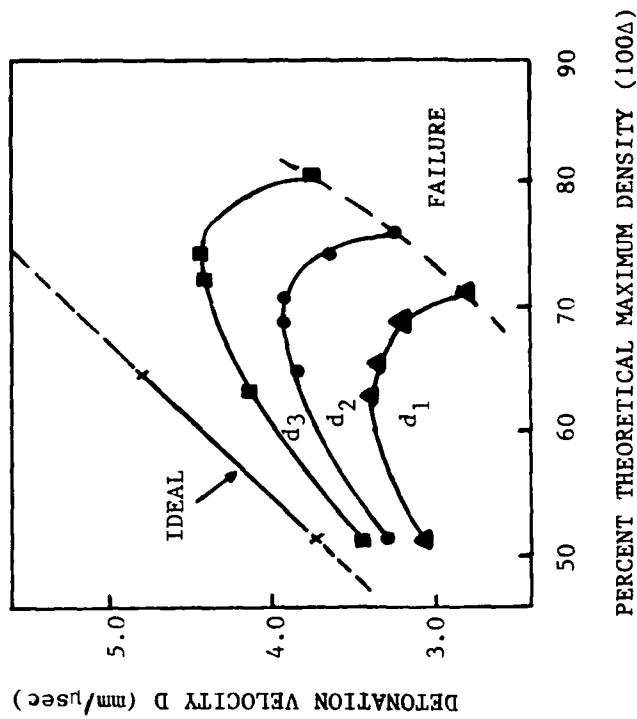


(b) Group 2 - Example shown is Dynammon (AN/peat meal, 88/12)

Figure 3. Shift of detonability limits caused by grinding the material (ref 5)



(a) Group 1 - Example shown is HBX-1 (RDX/TNT/Al/Wax, 40/38/17/5). The charge diameters d_1 to d_4 are 6.4, 12.7, 25.4, and 50.8 mm, respectively.



(b) Group 2 - Example shown is AP (average particle size 10 μ). The charge diameters d_1 to d_4 are 34.9, 50.8, and 76.2 mm, respectively.

Figure 4. Pattern of detonation velocity versus porosity curves at various charge diameters (ref 5)

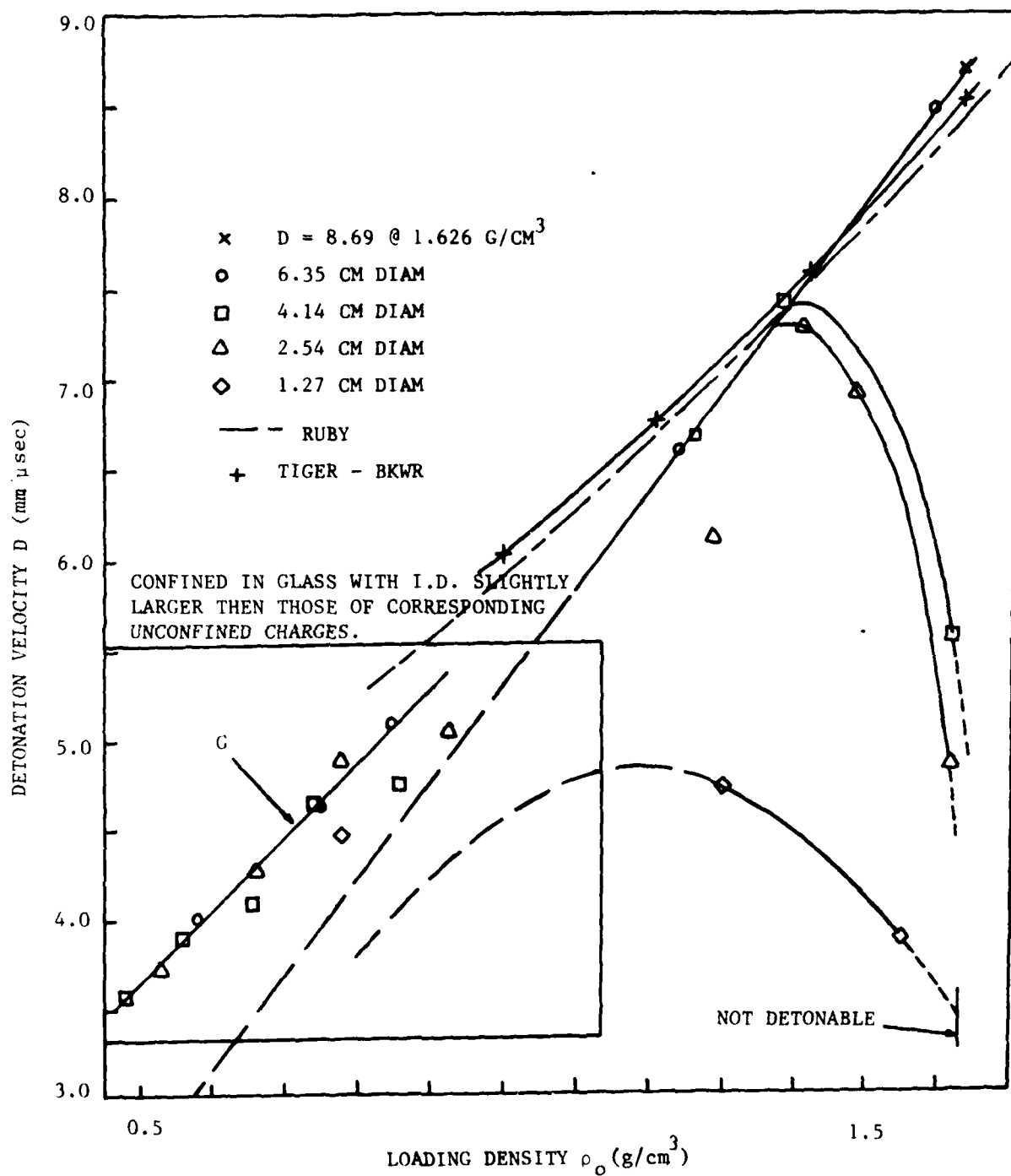


Figure 5. Detonation velocity of HN as a function of charge density and diameter (ref 19)

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